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Formation of short-lived radionuclides in the protoplanetary disk during late-stage irradiation of a volatile-rich reservoir

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The origin of short-lived ($t_{1/2}$ < 5 Myr) and now extinct radionuclides (^{10}Be, ^{26}Al, ^{36}Cl, ^{41}Ca, ^{53}Mn, ^{60}Fe; hereafter SLRs) is fundamental to understanding the formation of the early solar system. Two distinct classes of models have been proposed to explain the origin of SLRs[1–6]: (1) injection from a nearby stellar source (e.g., supernova, asymptotic giant branch star or Wolf-Rayet star) and (2) solar energetic particle irradiation of dust and gas near the proto-Sun. Recent studies have demonstrated that ^{36}Cl was extant in the early solar system[7–9]. However, its presence, initial abundance and the noticeable decoupling from ^{26}Al raise serious questions about the origin of SLRs. Here we report ^{36}Cl–^{36}S and ^{26}Al–^{26}Mg systematics for wadalite and grossular, secondary minerals in a calcium-aluminum-rich inclusion (CAI) from the CV chondrite Allende that allow us to reassess the origin of SLRs. The inferred abundance of ^{36}Cl in wadalite, corresponding to a ^{36}Cl/^{35}Cl ratio of $(1.81\pm0.13)\times10^{-5}$, is the highest ^{36}Cl abundance reported in any early solar system material. The high level of ^{36}Cl in wadalite and the absence of ^{26}Al (^{26}Al/^{27}Al $\leq 3.9\times10^{-6}$) in co-existing grossular indicates that (1) ^{36}Cl formed by late-stage solar energetic particle irradiation and (2) the production of ^{36}Cl, recorded by secondary minerals, is unrelated to the origin of ^{26}Al and other SLRs (^{10}Be, ^{53}Mn) recorded by primary minerals of CAIs and chondrules. We conclude that ^{36}Cl was produced by solar energetic particle irradiation of a volatile-rich reservoir in an optically thin protoplanetary disk adjacent to the accretion region of the CV chondrite parent asteroid.

Short-lived radionuclides provide a unique source of information about the astrophysical environment in which the solar system formed as well as high-resolution chronology of early solar system events[1–6]. The origin of SLRs in the early solar system, however, remains controversial[1]. Two main classes of models proposed – injection of SLRs from a stellar source and solar energetic particle (SEP) irradiation – have widely different consequences for the expected occurrences and distribution of SLRs in the early solar system. SLRs produced by stellar nucleosynthesis and injected into the protosolar molecular cloud are expected to homogenize quickly in the solar nebula; as a result, variations in their relative abundances may be ascribed to the passage of time[1,10]. In contrast, SLRs produced by SEP irradiation are more likely to be heterogeneously distributed, and variations in their relative abundances would reflect the local energetic particle environment[1].

Recently, excesses of ^{36}S correlated with $^{35}Cl/^{34}S$ ratios, were reported in sodalite ($Na_8Al_6Si_6O_{24}Cl_2$), a secondary mineral in CAIs and chondrules from the Allende (CV) and Ningqiang (CV anomalous) carbonaceous chondrites[7–9]. These studies provide clear evidence for *in situ* decay of ^{36}Cl in the early solar system; they also highlight the long standing dichotomy between the two classes of models proposed to explain the origin of SLRs. Theoretical calculations of SLR production by SEP irradiation[5,6], show that the highest reported ^{36}Cl (i.e., $^{36}Cl/^{35}Cl \sim 5\times10^{-6}$) levels in sodalite[7,8] are consistent with levels predicted for energetic particle irradiation of a reservoir with solar composition, but exceed by several orders of magnitude the values predicted for any stellar source[1]. This is consistent with astronomical observations of pre-main-sequence, solar-type stars that show powerful X-ray flares believed to be accompanied by intense fluxes of accelerated particles[11]. The irradiation models predict that the production of ^{36}Cl by SEP irradiation would not occur in isolation but be coupled to the production of other SLRs such as ^{26}Al, ^{53}Mn, and ^{10}Be[4–6]. However, this prediction is inconsistent with the absence of ^{26}Al in sodalite containing large ^{36}S excesses[7–9]. Moreover, high-precision ^{26}Al-^{26}Mg systematics of primary phases in chondrules from unmetamorphosed chondrites suggest that Earth, CAIs, and chondrules all formed from a reservoir with a homogeneous distribution of ^{26}Al, supporting its stellar origin[12]. These conflicting data underscore the importance of ^{36}Cl and its relationship to ^{26}Al for understanding the origin of SLRs in the early solar system. This issue can be resolved if we understand *when, where* and *how* ^{36}Cl formed and was incorporated into primitive meteorites containing a wide spectrum of objects – CAIs, chondrules, and matrix.

To address this problem, we studied a coarse-grained igneous CAI AJEF from the Allende meteorite. Primary minerals in AJEF (anorthite, melilite, pyroxene, and spinel) define an internal ^{26}Al–^{26}Mg isochron with an initial $^{26}Al/^{27}Al$ ratio of $\sim5\times10^{-5}$ (ref. 13). The melilite and anorthite are replaced by the co-existing secondary minerals, wadalite ($Ca_6(Al,Si,Mg)_7O_{16}Cl_3$) and grossular ($Ca_3Al_2Si_3O_{12}$) (Fig. 1; Supplementary Information (SI)), in which ^{36}Cl–^{36}S and

^{26}Al–^{26}Mg isotope systematics could be measured. Wadalite is a chlorine-rich mineral recently described in the Allende CAIs[14]. The high chlorine (~12 wt%) and very low sulfur content (<<0.01 wt%) make wadalite ideal for studies of the ^{36}Cl–^{36}S system.

The isotope abundances of chlorine and sulfur in wadalite in AJEF were determined using the Lawrence Livermore National Laboratory *Cameca* NanoSIMS 50 (see SI), a secondary ion mass spectrometer (SIMS) with nanometer scale spatial resolution. The isotope abundances of magnesium in grossular coexisting with wadalite were measured using the *Cameca* ims 1280 at the University of Hawai‘i (see SI).

The AJEF wadalite shows extremely large ^{36}S excesses with $^{36}S/^{34}S$ ratios of up to ~264 times that of the Canyon Diablo troilite standard value, correlated with the respective $^{35}Cl/^{34}S$ ratios (as high as $\sim 2\times10^{6}$; SI). The slope of a line fitted to the data yields an inferred $^{36}Cl/^{35}Cl$ ratio at the time of wadalite formation of $(1.81\pm0.13)\times10^{-5}$ (Fig. 2a). This value represents the highest initial abundance of ^{36}Cl reported in any meteorite and is more than four times greater than the highest $^{36}Cl/^{35}Cl$ initial ratio observed in sodalite in CAIs and chondrules[7–9].

Grossular associated with wadalite shows no resolvable ^{26}Mg excess (SI). The upper limit to the initial $^{26}Al/^{27}Al$ ratio in grossular is 3.9×10^{-6} (Fig. 2b). This value is similar to the upper limit obtained for sodalite in CV CAIs and chondrules[7–9]. The absence of radiogenic ^{26}Mg in secondary grossular contrasts with the well-constrained primary mineral internal isochron in AJEF[13] yielding an initial $^{26}Al/^{27}Al$ ratio of $\sim 5\times10^{-5}$ and suggests that the wadalite-grossular paragenesis formed >2.6 Myr after crystallization of the CAI.

The well-defined ^{26}Al-^{26}Mg chronologies, for both primary and secondary minerals, within AJEF place important constraints on the origin of ^{36}Cl. If ^{36}Cl was produced together with ^{26}Al at the birth of the solar system, the late formation of wadalite inferred from the low $(^{26}Al/^{27}Al)_0$ ratio in co-genetic grossular, would require an unrealistically high initial $^{36}Cl/^{35}Cl$ ratio of $>8.7\times10^{-3}$ at the time the primary CAI mineral assemblage crystallized. This value is more than sixty times the maximum level that can be produced by SEP irradiation of gas and/or dust of solar composition[5,6]. The high initial $^{36}Cl/^{35}Cl$ ratio in AJEF (1.8×10^{-5}) thus indicates that production of ^{36}Cl by SEP irradiation must have occurred late, >2 Myr after the formation of the first solar system solids and provides the first conclusive evidence that the ^{36}Cl found in secondary, low temperature minerals in CAIs and chondrules was produced in processes unrelated to those responsible for the SLRs (^{26}Al, ^{41}Ca, ^{10}Be) observed in primary, high temperature minerals in the same objects.

The suggestion that ^{36}Cl is produced by late-stage SEP irradiation must be evaluated against the predicted abundances of other SLRs, and compared to the observed abundances in early solar system solids. Evidence for late-stage production of SLRs, should be preserved in differentiated meteorites or in whole-rock samples of unequilibrated ordinary or carbonaceous chondrites. Assuming *late-stage* irradiation of a reservoir with solar composition and a particle fluence sufficient to produce ^{36}Cl corresponding to the inferred ^{36}Cl/^{35}Cl ratio in wadalite (~2×10^{-5}), we estimate, using the most recent calculations[5,6,15], relative abundances of the co-produced ^{26}Al, ^{53}Mn and ^{10}Be (Fig. 3). The range in predicted abundances reflects different assumptions among the models regarding production cross-sections, ^{3}He/H and ^{3}He/^{4}He ratios of SEP, hardness of the energy spectrum, and the relative importance of gradual to impulsive SEP events. The abundances of the three SLRs predicted here are compared against observed abundances in bulk meteorites[16–19] (Fig.3).

In nearly all cases, the amounts of ^{26}Al and ^{53}Mn produced by SEP irradiation of a solar composition reservoir are significantly greater than the values observed in bulk meteorites[16–19], and an irradiation model accounting for ^{36}Cl, ^{26}Al and ^{53}Mn in a self-consistent manner is difficult to achieve (Fig. 3). Only in the case of an extremely hard energy spectrum ($p \geq 5$) is a self-consistent solution achievable (Fig. 3). If the initial ^{36}Cl abundance, however, was any higher than the assumed value (i.e., ^{36}Cl/^{35}Cl > 2×10^{-5}), the problem will be exacerbated. The calculations presented above do not consider any delay between production and delivery of newly synthesized ^{36}Cl to the parent body or dilution of the irradiated product with unirradiated material. The ^{36}Cl abundance assumed for the calculation-predicted SLR abundances is likely a lower limit for the amount produced by late irradiation. Thus, ^{36}Cl production by late-stage SEP irradiation of a reservoir with solar composition would very likely overproduce both ^{26}Al and ^{53}Mn.

Overproduction of ^{26}Al and ^{53}Mn can be avoided if the reservoir irradiated to produce ^{36}Cl was depleted in refractory elements (enriched in volatile elements) relative to a solar composition due to CAI and chondrule formation. In particular, irradiation of a reservoir enriched in chlorine – a primary target element for SEP production of ^{36}Cl – would significantly enhance the production of ^{36}Cl relative to ^{26}Al and ^{53}Mn. During the lifetime of the protoplanetary disk, chlorine is present mainly as HCl gas[20]. It will condense as solid HCl hydrates ($HCl \cdot 3H_2O$) when temperatures fall below ~160 K and may adhere to mineral grains and water ice particles[20]. Solar energetic particle irradiation of either an HCl-rich gas or dust particles mantled by HCl hydrates would significantly enhance the production of ^{36}Cl relative to ^{26}Al and ^{53}Mn.

As oxygen is the main target element to produce ^{10}Be, ^{10}Be will be co-produced with ^{36}Cl in any late SEP irradiation scenario. However, late addition of ^{10}Be is difficult to detect in bulk meteorites because the small amount of ^{10}B produced by decay of ^{10}Be is likely to be overwhelmed by non-radiogenic ^{10}B (the solar B/Be ratio is ~2). The most sensitive test for the late addition of ^{10}Be is determination of boron-isotope abundances in late-forming secondary phases in CAIs or chondrules (e.g., wadalite or grossular). These measurements have not yet been performed but on the basis of the model presented here, we predict $^{10}Be/^{9}Be$ ratios exceeding 10^{-4} will be found.

The short half-life of ^{36}Cl requires that ^{36}Cl was incorporated into wadalite within 9×10^5 years (i.e., within three half-lives of ^{36}Cl) following production. This temporal constraint places limits on the location of SEP irradiation in the protoplanetary disk. Most models of SLR production by SEP irradiation assume the irradiation occurs near the co-rotation point of the Sun and the protoplanetary disk, known as the X-point[3], during the earliest stages of the solar system evolution when the Sun was a young (class 0) or accreting (class I) protostar. However, radial transport of material in the *latter stages* of the protoplanetary disk (when ^{36}Cl was produced) is inefficient[21] and we infer that formation of ^{36}Cl must have occurred adjacent to the region in which the CV chondrite parent asteroid accreted.

We thus propose that ^{36}Cl was largely produced by late-stage SEP irradiation of a volatile-rich reservoir in an optically thin protoplanetary disk while the Sun was a weak T Tauri star. Subsequently, ^{36}Cl accreted into the CV chondrite asteroid together with condensed water ices and was incorporated into secondary, chlorine-rich minerals, wadalite and sodalite, during prolonged parent body alteration. Delivery of chlorine as a component of water ice is consistent with the positive correlation between the chlorine content in chondrites and the degree of aqueous alteration[20].

Supplementary Information: Includes mineralogy and petrography of wadalite-grossular mineral assemblage in the Allende Type B CAIs, analytical methods, additional figures (S1-S4), and data tables (S1-S2) accompanies the paper.

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Figure captions:

Figure 1. Combined x-ray elemental map in Mg (red), Ca (green), and Al (blue) (a) and backscattered electron images (b, c) of the wadalite-grossular paragenesis in the Allende Type B CAI AJEF. Regions outlined in panels (a) and (b) are shown in greater detail in panels (b) and (c), respectively. Wadalite together with grossular and monticellite occurs in secondary veins crosscutting primary melilite. an = anorthite; grs = grossular; mel = melilite; mnl = monticellite; sp = spinel; wdl = wadalite.

Figure 2. Panel (a): ^{36}Cl-^{36}S isochron diagram of wadalite from the Allende CAI AJEF. The solid line represents a weighted, least-squares regression through the data and corresponds to $(^{36}Cl/^{35}Cl)_0 = (1.81\pm0.13)\times10^{-5}$. Stippled lines represent the error envelope. The dashed line and lower slope in the inset represents the inferred $(^{36}Cl/^{35}Cl)_0$ ratio for sodalite from the Allende CAI Pink Angel[2]. Panel (b): ^{26}Al-^{26}Mg isochron diagram for grossular in the Allende CAI AJEF. The black solid line represents a weighted, least-squares regression through the data corresponding to $(^{26}Al/^{27}Al)_0 = (1.1\pm2.8)\times10^{-6}$. Dashed lines represent the error envelope. The uncertainties in both panels (a) and (b) and inset are 2σ.

Figure 3. Ratio of calculated to observed abundances of ^{10}Be, ^{26}Al, and ^{53}Mn assuming a particle fluence sufficient to produce ^{36}Cl corresponding to $(^{36}Cl/^{35}Cl) = 2\times10^{-5}$. The calculated ^{26}Al and ^{53}Mn abundances are normalized to the inferred upper limits of ^{26}Al and ^{53}Mn abundances for bulk meteorite samples corresponding to $^{26}Al/^{27}Al \leq 6\times10^{-6}$ and $^{53}Mn/^{55}Mn \leq 9\times10^{-6}$, respectively. There are no constraints on the ^{10}Be abundance in bulk meteorites; instead the calculated abundance for ^{10}Be is normalized to the inferred solar initial value (i.e., $^{10}Be/^{9}Be = 1\times10^{-3}$). The range of ratios reflects different assumptions among the models regarding the production cross-sections, $^{3}He/H$ and $^{3}He/^{4}He$ ratios of the SEP, the hardness of the energy spectrum and the relative importance of gradual to impulsive SEP events.

Figures:

Fig. 1

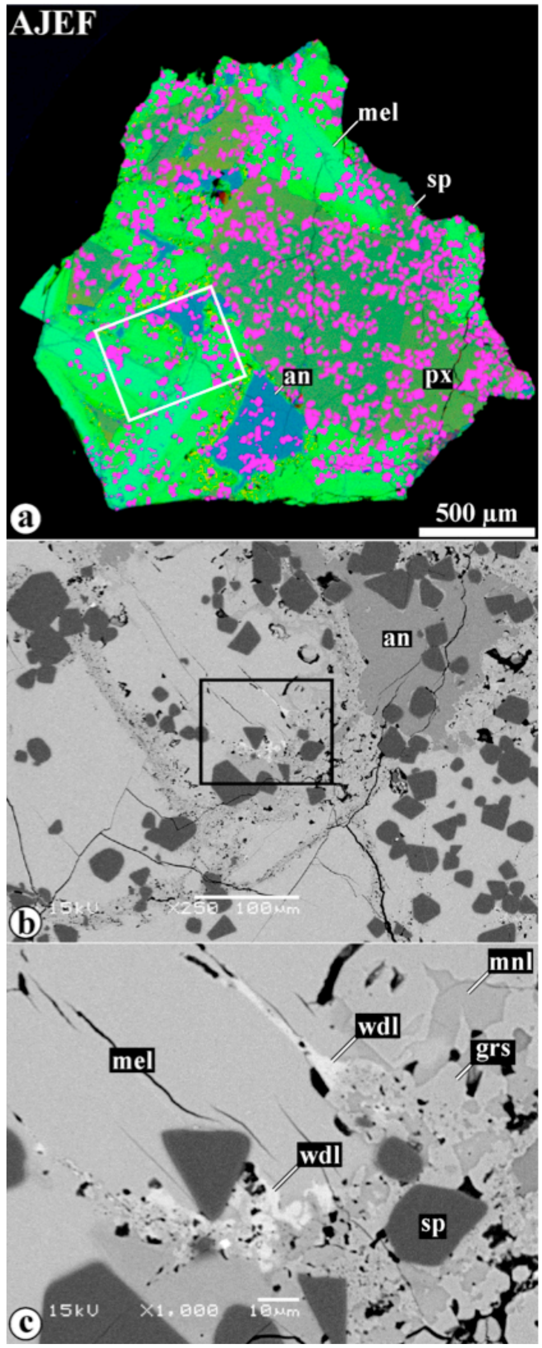
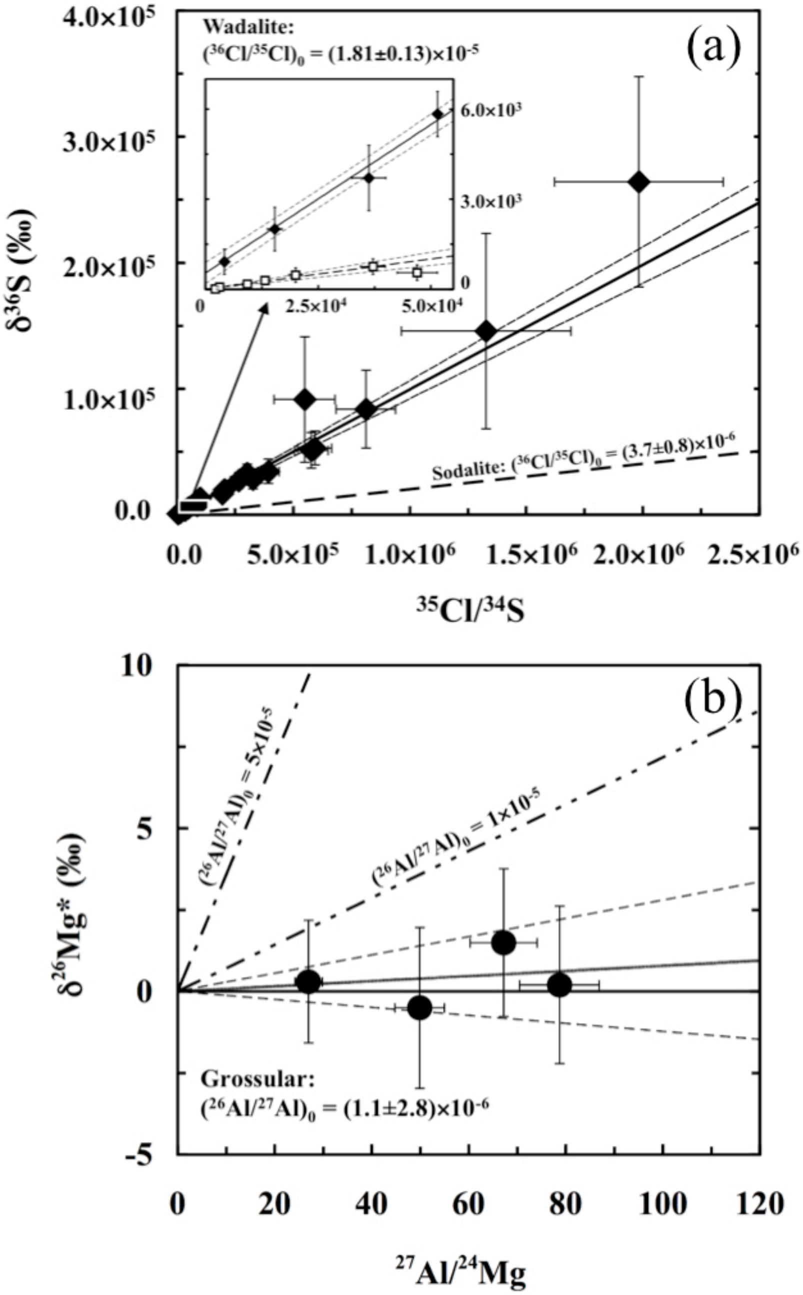
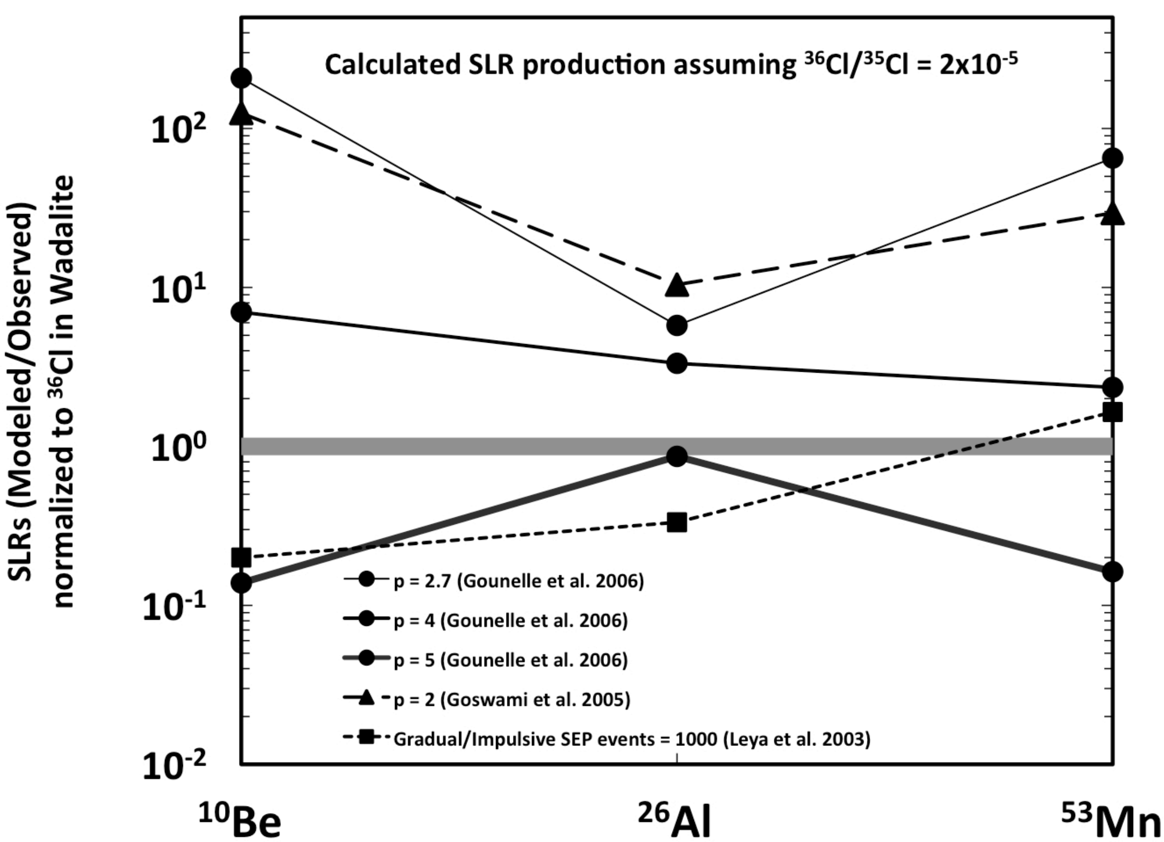


Fig. 2



(a)
Wadalite:
(^{36}Cl/^{35}Cl)$_0$ = (1.81±0.13)×10^{-5}
Sodalite: (^{36}Cl/^{35}Cl)$_0$ = (3.7±0.8)×10^{-6}
δ^{36}S (‰)
^{35}Cl/^{34}S
(b)
(^{26}Al/^{27}Al)$_0$ = 5×10^{-5}
(^{26}Al/^{27}Al)$_0$ = 1×10^{-5}
Grossular:
(^{26}Al/^{27}Al)$_0$ = (1.1±2.8)×10^{-6}
δ^{26}Mg* (‰)
^{27}Al/^{24}Mg

Fig. 3



Calculated SLR production assuming 36Cl/35Cl = 2x10-5
SLRs (Modeled/Observed) normalized to 36Cl in Wadalite
10^2
10^1
10^0
10^-1
10^-2
10Be
26Al
53Mn
p = 2.7 (Gounelle et al. 2006)
p = 4 (Gounelle et al. 2006)
p = 5 (Gounelle et al. 2006)
p = 2 (Goswami et al. 2005)
Gradual/Impulsive SEP events = 1000 (Leya et al. 2003)